AMENDMENT (2)

(Amendment Based on Article 11)

To: The Commissioner of Japanese Patent Office

1. Identification of the International Application PCT/JP2004/000007

2. Applicant
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4. Items to be amended: Specification and Claims

5. Content of Amendment
   (1) The expression "by heating a sol containing a metal or semimetal alkoxide, and a polyorganosiloxane having functional group(s) reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000, plus a highly thermally conductive filler, to gel said sol" on page 2 line 12 to 16 in Specification should be amended as "by coating a sol containing a metal or semimetal alkoxide, a polyorganosiloxane having functional group(s) reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000, and a highly thermally conductive filler, on a base and heating said sol, to gel said sol"
   (2) The expression "Further, it is preferable that said organosilicon

compound is a polyorganosiloxane having functional group(s) that are reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being in the range of between 400 and 15000, or said organosilicon compound is a polyorganosiloxane having functional group(s) that are reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000 in a case where special heat resistance is required." on page 2 lines 20 to 28 in Specification should be deleted.

(3) The expression "Generally, polyoriganosiloxane having a weight average molecular weight is in the range between of 400 and 80000 is used in the present invention, and considering heat resistance, polyorganosiloxane having a weight average molecular weight of higher than 15000 is preferable. In a case where molded organic-inorganic hybrid is used under the conditions at a low temperature of below 200°C, polyorganosiloxane having a weight average molecular weight in the range between of 400 and 15000 is preferably used. Under temperature conditions higher than 200°C, polyorganosiloxane, having a weight average molecular weight in the range of 15000 and 80000 is preferably used." on page 4 lines 25-34 in Specification should be amended as "Considering heat resistance, generally polyorganosiloxane having a weight average molecular weight of higher than 15000 is used. In a case where molded organic-inorganic hybrid is used, under temperature conditions higher than 200°C, polyorganosiloxane, having a weight average molecular weight in the range of 15000 and 80000 is preferably used".

(4) The expression "heating a sol containing a metal or semimetal alkoxide, and a polyorganosiloxane having functional group(s) reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000, plus a highly thermally conductive filler, to gel said sol" on page 15 in Claim 1

should be amended as "coating a sol containing a metal or semimetal alkoxide, a polyorganosiloxane having functional group(s) reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000, and a highly thermally conductive filler, on a base and heating said sol, to gel said sol.".

6. The list of attached paper: Specification page 2 and 4
Claims on page 15

resistance to enable ON, and energy saving, said roller having a structure consisting of a base made of a silicon rubber to which a filler is added, and a surface layer made of fluorocarbon resin. As for said roller, there is a problem in that the amount of filler that can be added to said silicon rubber is limited, and, as a result, high thermal conductivity cannot be realized, extending the time it takes to heat said roller to a predetermined temperature, to retard instant ON.

DISCLOSURE OF THE INVENTION

To solve these problems, the present invention provides a heat-resistant, thermally conductive material being made from an organic-inorganic hybrid material, prepared by coating a sol containing a metal or semimetal alkoxide, a polyorganosiloxane having functional group(s) reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000, and a highly thermally conductive filler, on a base and heating said sol, to gel said sol.

Said organosilicon compound is preferably organosiloxane having functional group(s) that are reactive with said metal or semimetal alkoxide at one or both ends.

Further, it is preferable that said organic-inorganic hybrid material is synthesized by the condensation reaction between the reactive functional group(s) at one or both ends of said organosilicon compound and said metal or semimetal alkoxide, accompanying hydrolysis, and said condensation reaction is preferably carried out by heating at a temperature higher than 80°C to decrease the viscosity of said organosilicon compound.

Further, it is also preferable that the metal of said metal alkoxide is of one or more kind(s) of metal(s) selected from a group consisting of boron aluminum,

The metal or semimetal of metal or semimetal alkoxide used in the present invention is such as aluminium, silicon, titanium, vanadium, manganese, iron, cobalt, zinc, germanium, yttrium, zirconium, niobium, lanthanum, cerium, cadmium, tantalum, and tungsten, or the like, said metals or semimetals being able to produce alkoxide. In particular, preferable metals may be such as titanium, zirconium, and silicon.

Any kind of alkoxide such as methoxide, ethoxide, propoxide, butoxide or the like may be used in the present invention, and said metal or semimetal alkoxide preferably being chemically modified with a chemical modifier such as acetoacetate, such as methyl acetoacetate, ethyl acetoacetate, isopropyl acetoacetate or the like.

(Organosilicon compound)

As said organosilicon compound of the present invention, such as dialkyl dialkoxysilane, preferably polyorganosiloxane, having functional group(s) at one or both ends reactive with said metal or semimetal alkoxide such as polydimethyl siloxane having a silanol group at one or both end may be used. Said dialkyldialkoxysilane may be such as dimethyldimethoxysilane, dimethyldiethoxysilane, dimethyldipropoxysilane, dimethyldibutoxysilane, diethyldimethoxysilane, diethyldiethoxysilane, diethyldipropoxysilane, diethyldibutoxysilane, dipropyldimethoxysilane, dipropyldiethoxysilane, dipropyldipropoxysilane, dipropyldibutoxysilane, diphenyldimethoxysilane, diphenyldiethoxysilane, diphenyldipropoxysilane, diphenyldibutoxysilane and the like.

Considering heat resistance, generally polyorganosiloxane having a weight average molecular weight of higher than 15000 is used. In a case where molded organic-inorganic hybrid is used, under temperature conditions higher than 200°C, polyorganosiloxane, having a weight average molecular weight in the range of 15000 and 80000 is preferably used.

CLAIMS

1. (Amended) A heat-resistant, thermally conductive material being made from an organic-inorganic hybrid material, prepared by heating a sol containing a metal or semimetal alkoxide, and a polyorganosiloxane having functional group(s) reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000, plus a highly thermally conductive filler, to gel said sol.
2. (Deleted)
3. (Deleted)
4. (Deleted)
5. (Amended) A heat-resistant, thermally conductive material in accordance with Claim 1, wherein said organic-inorganic hybrid material is synthesized by the condensation reaction between the reactive functional group(s) at one or both ends of said organosilicon compound and said metal or semimetal alkoxide, accompanying hydrolysis.
6. (Amended) A heat-resistant, thermally conductive material in accordance with Claims 1 and 2, wherein the metal of said metal alkoxide is of one or more kind(s) of metal(s) selected from a group consisting of boron aluminum, silicon, titanium, vanadium, manganese, iron, cobalt, zinc, germanium, yttrium, zirconium, niobium, lanthanum, cerium, cadmium, tantalum and tungsten.
7. (Amended) A heat-resistant, thermally conductive material in accordance with Claims 1 to 3, wherein said highly thermally conductive filler is a fine powder of one or more kind(s) of metal and/or metal oxide and/or metal nitride and/or metal carbide.

AMENDMENT (1)

(Amendment Based on Article 11)

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4. Items to be amended: Specification and Claims

5. Content of Amendment
   (1) The expression "an organosilicon compound" on page 2 line 13 in Specification should be amended as "a polyorganosiloxane having functional group(s) reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000"
   (2) The expression "an organosilicon compound" on page 15 line 5 in Claim 1 should be amended as "a polyorganosiloxane having functional group(s) reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000".
   (3) Claims 2 to 4 on page 15 should be deleted.
   (4) The expression "Claims 1 to 4" on page 15 in Claim 5 should be amended as "Claim 1".

(5) The expression "Claims 1 to 5" on page 15 in Claim 6 should be amended as "Claims 1 and 2".

(6) The expression "Claims 1" on page 15 in Claim 7 should be amended as "Claims 1 to 3".

6. The list of attached paper: Specification page 2 and 2/1

Claims on page 15

CLAIMS

1. (Amended) A heat-resistant, thermally conductive material being made from an organic-inorganic hybrid material, prepared by heating a sol containing a metal or semimetal alkoxide, and a polyorganosiloxane having functional group(s) reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000, plus a highly thermally conductive filler, to gel said sol.
2. (Deleted)
3. (Deleted)
4. (Deleted)
5. (Amended) A heat-resistant, thermally conductive material in accordance with Claim 1, wherein said organic-inorganic hybrid material is synthesized by the condensation reaction between the reactive functional group(s) at one or both ends of said organosilicon compound and said metal or semimetal alkoxide, accompanying hydrolysis.
6. (Amended) A heat-resistant, thermally conductive material in accordance with Claims 1 and 2, wherein the metal of said metal alkoxide is of one or more kind(s) of metal(s) selected from a group consisting of boron aluminum, silicon, titanium, vanadium, manganese, iron, cobalt, zinc, germanium, yttrium, zirconium, niobium, lanthanum, cerium, cadmium, tantalum and tungsten.
7. (Amended) A heat-resistant, thermally conductive material in accordance with Claims 1 to 3, wherein said highly thermally conductive filler is a fine powder of one or more kind(s) of metal and/or metal oxide and/or metal nitride and/or metal carbide.

REPLY (1)

To: The Commissioner of Japanese Patent Office

1. Identification of the International Application PCT/JP2004/000007

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4. Date of Notification: March 16, 2004

5. Content of Reply
   Claim 1 is amended as follows.
   1. A heat-resistant thermally conductive material being made from an organic-inorganic hybrid material, prepared by heating a sol containing a metal or semimetal alkoxide, a polyorganosiloxane having functional group(s) reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000, and a highly thermally conductive filler, to gel said sol.

   In a case where the weight average molecular weight of said polyorganosiloxane which composes the organic part of said organic-inorganic hybrid material is beyond 15000, the heat resistance of said organic-inorganic hybrid materials, improves, so that said organic-inorganic hybrid material can be used at temperature conditions

higher than 200°C. Further, since the mechanical strength of said organic-inorganic hybrid material improves, even if said highly thermally conductive filler is added to said organic-inorganic hybrid material in an increased amount, the mechanical strength of said organic-inorganic hybrid material is not degraded.

Accordingly, said organic-inorganic hybrid material has high mechanical strength and thermal conductivity.

COMPARISON AND CONSIDERATION

(1) Literature 1 (JP10-245490A)

Literature 1 discloses a particle dispersed inorganic-organic hybrid type elastomer, in which particles are dispersed in an elastomer synthesized from a metal alkoxide and silanol terminated polydimethyl siloxane (corresponding to said organic-inorganic hybrid material of the present invention), said particles including titanium oxide, aluminum oxide, silicon oxide, which are each used for said thermally conductive filler of the present invention.

Nevertheless, Literature 1 does not disclose that the weight average molecular weight of said silanol terminated polydimethylsiloxane, which composes the organic part of said inorganic-organic hybrid type elastomer is higher than 15000.

(2) Literature 2 (JP2000-38508 A)

Literature 2 discloses a polyorganosiloxane compound being curable at room temperature containing (A) a silicon functional polyorganosiloxane having two or more silicon functional groups, (B) a curing catalyst, (C) a thermally conductive filler, and (D) titanium dioxide.

Said (A) polyorganosiloxane is included in said polyorganosiloxane of the present invention and as said (C) thermally conductive filler, alumina powder (aluminum oxide), silicon carbonate, aluminum nitride, boron nitride, and the like, which are used as said highly thermally conductive filler, are disclosed.

However, (A) polyorganosiloxane does not react with a metal alkoxide to form an organic・inorganic hybrid material, but instead produces a cross-linking structure through a curing catalyst. Accordingly, it is not

disclosed in Literature 2 that polyorganosiloxane having a weight average molecular weight of higher than 15000 is used as an organic component of said organic-inorganic hybrid.

(3) Literature 3 (JP-278311A)

Literature 3 discloses an inorganic-organic fusion compound (inorganic-organic hybrid) in which inorganic particles are dispersed, said inorganic-organic fusion compound being prepared by reacting alkyl alkoxysilan and a metal or semimetal alkoxide. However, organic component used in said inorganic-organic fusion compound is alkylalkoxy silan not polyorganosiloxane.

(4) Literature 4 (JP 63-81176A)

Literature 4 discloses a compound in which said highly thermally conductive filler of the present invention is mixed into a reactant of organosiloxane and a metal alkoxide (organic-inorganic hybrid). The organic component in said organic-inorganic hybrid, however, is organosiloxane, and not polyorganosiloxane.

CONCLUSION

As described above, none of Literatures 1 to 4 disclose the present invention, and the effect of the present invention being that material having a high mechanical strength and thermal conductivity is obtainable, is unforeseen from Literatures 1 to 4.

6. The list of attached paper: Amendment

REPLY (2)

To: The Commissioner of Japanese Patent Office

1. Identification of the International Application PCT/JP2004/000007

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4. Date of Notification: December 21, 2004

5. Content of Reply
   Claim 1 is amended as follows.
   1. A heat-resistant thermally conductive material being made from an organic-inorganic hybrid material, prepared by coating a sol containing a metal or semimetal alkoxide, a polyorganosiloxane having functional group(s) reactive with said metal or semimetal alkoxide at one or both ends, with the weight average molecular weight of said polyorganosiloxane being higher than 15000, and a highly thermally conductive filler, on a base and heating said sol, to gel said sol.

   In the present invention relating to said amended Claim 1, said sol has a high viscosity, so that even if said sol is coated on said base to form a thin film, the result will be a uniformly smooth thin film of said organic-inorganic hybrid having a uniform thickness, and further, said sol

can be coated onto said base to form a thick film, and in this case a highly elastic thick film of said organic-inorganic hybrid can be obtained. Said thick film may adhere closely to said base(s) when said thick film is formed on a side of said base, or put between a pair of said bases. Further, said organic-inorganic hybrid material has excellent heat resistance. Since said organic-inorganic hybrid material is used as a thermally conductive material, which is often exposed to high temperature conditions, heat resistance is an important property for said organic-inorganic hybrid material.

Literature 1 (JP10-245490A) discloses a structure wherein an organic-inorganic hybrid type elastomer with dispersed particles is used, said structure being substantially the same as the structure of the present invention, and said particles including TiO_2, SiO_2, Al_2O_3, and the like, which are used as said highly thermally conductive filler of the present invention. However, since Literature 1 does not disclose that the weight average molecular weight of the polydimethylsiloxane is to be higher than 1500, the above described effect of the present invention, as it relates to coating workability, is unforeseen from the disclosure of Literature 1.

Literature 2 (JP2000-38508) discloses a polyorganosiloxane, and since a semimetal alkoxide such as tetramethoxysilane is used as a cross-linking agent for said polyorganosiloxane, said polyorganosiloxane corresponds with said organic-inorganic hybrid of the present invention, and TiO_2 (fumous_titaniumoxide) used as said highly thermally conductive filler in the present invention is also added to said polyorganosiloxane. However, since Literature 2 does not disclose that the weight average molecular weight of said polyorganosiloxane is set to be higher than 15000 in the present invention, the above described effect of the present invention, as it relates to coating workability, is unforeseen from the disclosure of Literature 2.

Literature 3 (H07-278311A) discloses an organic-inorganic fusing compound which uses alkylalkoxysilane as an organic composition. Accordingly, since said polyorganosiloxane, having a weight average molecular weight of higher than 15000, is not used, the above described effect of the present invention, as it relates to coating workability, is

unforeseen from the disclosure of Literature 3.

Literature 4 (S63-81176A) discloses a compound for coating which uses an organoalkoxysilane, the same as in Literature 3, and a polyorganosiloxane is not used in Literature 4. Accordingly, Literature 4 does not disclose that the weight average molecular weight of said polyorganosiloxane is set to be higher than 15000 in the present invention, so that the above described effect of the present invention, as it relates to coating workability, is unforeseen from the disclosure of Literature 4.

Literature 5 (H11-130864) discloses an organic-inorganic hybrid material in which the inorganic particles are dispersed, said inorganic particles including such as Al_2O_3, TiO_2, SiO_2, and the like, with polyorganosiloxane being also used in the organic composition. Literature 5, however, does not disclose that the weight average molecular weight of said polyorganosiloxane is set to be higher than 15000 in the present invention, so that the above described effect of the present invention, as it relates to coating workability, is unforeseen from the disclosure of Literature 5.

As described above, in the present invention, since the above described unforeseen effect is realized through above described limitation of molecular weight of polyorganosiloxane, the present invention can not be easily thought of by the expert referring to the disclosures of Literatures 1 to 5.